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16. ABSTRACT

The paper gives an overview of stationary source NOx control technology. Fuel combustion is the major source of NOx emissions in populated areas. In the U.S., stationary combustion sources account for 53% of the NOx produced by human activity; industrial non-combustion sources produce about 2%. The rest is produced by mobile sources. Stationary source emissions can be controlled by modifying the combustion process or by post-combustion treatment. Conventional combustion modification can reduce NOx by up to 50%; advanced techniques have the potential for over 80% reduction. Post-combustion treatment can achieve up to 90% control; however, costs may be substantially higher than those for combustion modification.

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Stationary Source NO_x Control Technology Overview

By F. T. Princiotta

ABSTRACT

The major source of NO_x emissions in populated areas is combustion of fuels. In the U. S., stationary combustion sources account for 53 percent of the NO_x produced by human activity, while industrial non-combustion sources produce about 2 percent. The balance is produced by mobile sources. Stationary source emissions can be controlled by modification of the combustion process or by post-combustion treatment. Conventional combustion modification techniques can achieve up to 50 percent reduction of NO_x, while advanced techniques have the potential for over 80 percent reduction. Post-combustion treatment techniques can achieve up to 90 percent control; however, costs may be substantially higher than those for the combustion modification techniques.

INTRODUCTION

Nitrogen oxides (NO_x) is a class of pollutants which has received widespread attention over the last 25 to 30 years. The principal species of concern are nitric oxide (NO) and nitrogen dioxide (NO_2). The NO_x produced by industrialized societies is concentrated in populated areas or in effluent streams which are transported over long distances. Most of the NO_x is produced as NO and then a portion is transformed into NO_2 by atmospheric processes.

ENVIRONMENTAL EFFECTS

The presence of NO_x in the atmosphere contributes to a number of potential environ-mental effects. These effects occur through transformation of NO_x by thermal processes or by reaction with other species. Specific examples include photochemical smog, health effects, and acid rain.

Photochemical Smog. One of the earliest concerns related to the role of NO_x is in the formation of photochemical smog. Reaction of NO_x with hydrocarbons in the presence of sunlight results in elevated levels of a number of oxidants, including ozone. These compounds result in reduced visibility, eye irritation, and other effects on people and vegetation.

Health Effects. While NO is relatively innocuous at atmospheric concentrations, respiratory effects can occur once it is transformed into NO_2. Studies have indicated effects from both long term exposure to low levels and short term exposure to higher levels. In the U. S., a long term exposure ambient NO_2 standard is in effect, and a short term standard is being studied.

Acid Rain. The most recent concern is the role of NO_x and SO_x as precursors for acidic species in rainfall. The theories relating acid rain to industrial pollution suggest two possible mechanisms: 1) conversion of NO_x and SO_x in power plant plumes with deposition some distance away from the source; and 2) local deposition of emissions of sources with shorter stacks.

SOURCES

In the United States, emissions of NO_x are divided approximately equally between stationary and mobile sources. Figure 1 (Ref. 1) illustrates the relative contributions of major sources. By contrast about 95 percent of the SO_x is from stationary sources. Both the sources of emissions and the trends must be considered in the development of control technology.

The distribution of NO_x by stationary source category is shown in Figure 2 (Ref. 1). Utility boilers produce 53 percent of the emissions, and industrial boilers produce another 14 percent. A large fraction of the boilers are fired with coal

and heavy fuel oil. The other major categories are stationary internal combustion and gas turbine engines (22 percent), industrial process furnaces (5 percent), and warm air furnaces (2 percent).

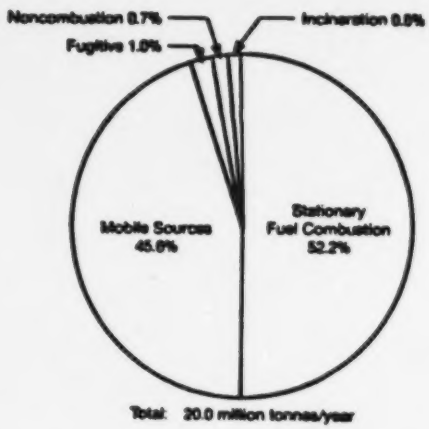

Figure 1. Distribution of human-produced NO_x emissions by major source.

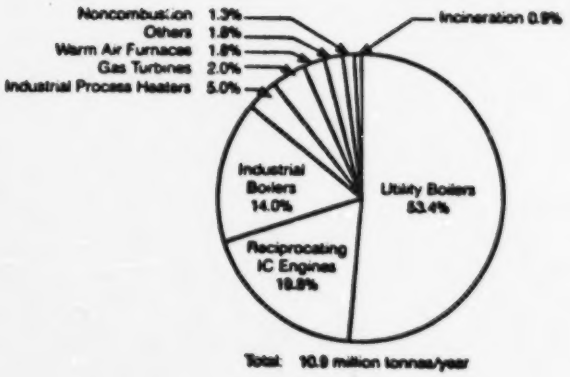

Figure 2. Distribution of U.S. NO_x emissions for 1980 by stationary source category.

Trends. The prediction of future trends in emissions from stationary sources is dependent on a number of factors, including fuel availability, energy use growth rates, emission control requirements, and economics. In general, the trends appear to be to greater NO_x emissions from coal combustion due to fuel switching and to lower relative mobile source contributions due to relatively stringent automobile emission regulations. Figure 3 shows what may be an upper limit projection for NO_x emissions in the U. S. The assumptions are: high energy consumption growth; low nuclear; high coal combustion and existing controls on mobile source emissions. Assuming no additional New Source Performance Standards (NSPS), the result is over a 100 percent increase in stationary source NO_x by 2000 but very little change in mobile source emissions. Note that more stringent standards (moderate and stringent NSPS cases) can substantially decrease NO_x emissions. By contrast, due to required control levels on new plants (i.e., 70-90 percent removal), nationwide SO_x emissions are relatively unchanged in the next two decades.

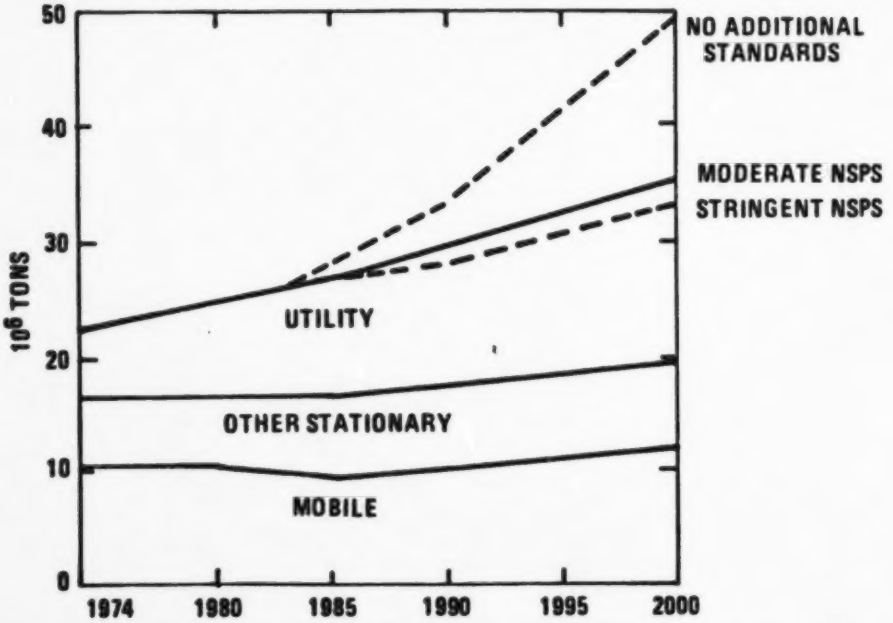

Figure 3. NO_x emissions projections high growth scenario (Ref. 2).

CONTROL TECHNOLOGY

The two general categories of control technology are combustion modification (CM) and post-combustion treatment processes. In the past 15 years considerable effort has been devoted to development and commercialization of various control technologies. CM has emerged as the technology which can give moderate to substantial NO_x reductions at a relatively low cost. Post-combustion treatment is normally employed in addition to CM where a high level of control is required even though higher capital and operating costs are incurred. The papers in this session will explore these techniques in detail; however, a brief description of each technology will provide an overall background.

Combustion Modification. NO_x is formed during combustion by two mechanisms: thermal NO_x is formed by fixation of molecular nitrogen from combustion air in high temperature regions of the flame zone; and fuel NO_x is formed by oxidation of nitrogen compounds chemically bound in the fuel molecule. CM for NO_x is based on altering the chemical and/or thermal conditions under which air/fuel reactions occur. Thermal NO_x can be controlled by reducing peak temperatures, while fuel NO_x can be controlled by reducing oxygen availability in the flame zone where the nitrogen is being evolved from the fuel molecules. The implementation of CM can be separated into two subclasses: 1) conventional technology which involves relatively minor changes in equipment and/or operating conditions; and 2) advanced techniques which involve more extensive changes to achieve more effective control.

A number of conventional techniques have been tested on existing systems and/ or fitted to the new boilers being sold. These techniques require modification of operating conditions or minor hardware changes. Although the NO_x reductions are moderate (typically 10 to 50 percent), the techniques are

low cost. The three most common techniques are flue gas recirculation, staged combustion, and first-generation low-NO_x burners.

1) Flue gas recirculation (FGR) provides for recirculation of relatively cool combustion gases from the stack and for mixing with the combustion air. This provides an inert diluent which decreases peak temperatures and reduces thermal NO_x. While NO_x reductions of up to 90 percent have been reported in experimental systems burning clean fuels, practical systems typically are limited to 50 percent by operational considerations. For nitrogen-containing fuels (e.g., many coals), FGR is much less effective, giving only 10-30 percent reductions in experimental and practical systems.

2) Staged combustion (SC) is a technique which reduces oxygen availability in the burner zone and provides air for complete combustion after the primary combustion zone. It has been implemented by modifying the air distribution between burners in multiple burner boilers or by the addition of ports above the active burner zone. SC not only reduces oxygen availability but also reduces peak flame temperature by delaying heat release. As a result it is effective for control of both thermal and fuel NO_x. In practical systems, NO_x reductions are from 20 to 50 percent. Although the technique has potential for greater reductions, it is limited by operational problems which occur if the burner zone is operated very fuel-rich.

3) First-generation low NO_x burners (LNB) is another method of delaying air/ fuel mixing thereby reducing both thermal and fuel NO_x. The burners are based on changes in fuel injector design and secondary air introduction method. A number of commercial low NO_x coal burners, in use on practical boilers, achieve NO_x reductions of 20-40 percent from uncontrolled levels. The approaches to, and results of, application of conventional modifications are discussed in later papers.

Advanced combustion modification is being developed using the same basic approaches to achieve more effective NO_x control and to maintain the cost advantages of conventional CM. The general approaches include second-generation low NO_x burners, prechamber burners, and novel concepts.

1) Second Generation Low-NO_x Burners. An advanced concept that is most readily adaptable to existing furnace designs is based on controlling air/fuel mixing aerodynamically by changes in burner design. This approach involves fuel and air introduction schemes which create a fuel-rich core in the flame for fuel NO_x control and a surrounding air-rich zone for carbon burnout. The distributed-mixing coal burner is an example of this approach. A promising technology utilizing low NO_x burners for both NO_x and SO_x removal is Limestone-Injection Multistage Burners (LIMB). Results both in the U. S. and the Federal Republic of Germany suggest that high levels of SO_x can be realized by the introduction of an alkali such as limestone either mixed with the coal or introduced through staged air ports. Such an approach could represent a breakthrough in achieving low cost SO_x/NO_x removal from existing and new coal-fired boilers.

2) Prechamber Burners. Considerably better control of combustion parameters can be achieved by providing a physical separation of the fuel-rich zone and the subsequent burnout zone. In this approach the thermal environment can also be better controlled.

3) Novel Concepts. For certain applications it is possible to abandon the diffusion flame entirely and use a new approach to fuel combustion. Catalytic combustion may be effective for gaseous fuels where mixing problems are minimal.

Post-Combustion Techniques. A number of techniques can destroy NO_x in the flue gas by reaction with a reagent (e.g., NH_3) or by physical absorption. The techniques are capable of NO_x removal of up to 90 percent; however, capital and operating costs are typically significantly higher than those for CM. The techniques can be used singly or in combination with CM where very high NO_x reduction is necessary.

NO_x can be reduced to N_2 by reactions with reagents that generate amine or hydrocarbon radicals. The most common reagents are ammonia (NH_3) and gaseous hydrocarbon fuels (e.g., methane). The reactions can occur in the gas phase (homogeneous) or in the presence of a catalyst (heterogeneous). In addition, the reactions can occur in the presence of oxygen (selective) or may require the absence of oxygen (non-selective). The post-combustion techniques provide various methods of contacting NO_x with the reagents under conditions necessary for the reduction reactions described below.

1) Non-selective reduction occurs when NO_x is contacted with a reagent under oxygen-deficient conditions. The reducing reagent is normally a fuel constituent (hydrocarbon, CO, or hydrogen) which is introduced into an oxygen-rich fuel stream or is present in the products of incomplete combustion. The reduction can be either homogeneous or hetrogeneous. The reducing zone is followed by an oxygen-rich zone to consume the balance of the reducing agent. If a supplemental fuel is required, provision for heat recovery may be necessary for efficiency reasons.

2) Selective homogeneous reduction involves reaction of NH_3 with NO_x at elevated temperatures (about 1000°C) in the presence of oxygen. In laboratory systems, up to 90 percent reductions have been reported. For practical systems, the NH_3 must be injected in the region near the end of the radiant furnace.

Provision must be made for injection locations in the proper temperature regime or for modification of the temperature at which the reaction takes place by hydrogen injection. In practice, due to mixing and temperature control problems, reductions have been restricted to about 50 percent.

3) Selective catalytic reduction allows reaction of NO_x with NH_3 at lower temperatures (150°C) under more controlled conditions. For gas-, oil-, and coal-fired experimental practical systems, NO_x reductions of up to 90 percent have been reported. Significant capital and operating costs are incurred.

SUMMARY

A wide variety of techniques are available to reduce NO_x emissions from stationary sources. Conventional combustion techniques offer NO_x reductions of up to 50 percent, while advanced techniques may achieve 80-90 percent from uncontrolled levels. Post-combustion treatment techniques also offer removals up to 90 percent; however, costs are generally significantly higher.

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